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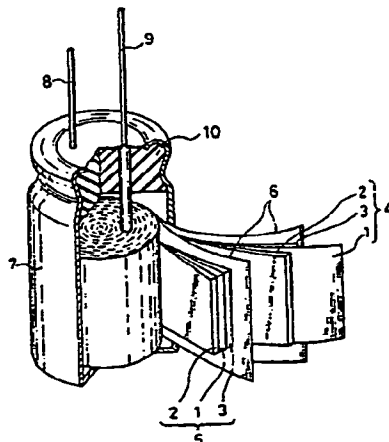
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(54) **Electric double layer capacitor and method for producing the same.**

(57) An electric double layer capacitor which comprises a pair of polarizable electrode, composed of electrically-conductive substrates coated with layers formed on said substrates, of a mixture of activated carbon with a water-soluble material-based binding agent, optionally including an agent for improving electric conductivity, facing with each other by being interposed with a separator, and an electrolyte is disclosed. The capacitor has both the advantageous features of the conventional ones of the aqueous electrolyte and of the organic solvent electrolyte. More particularly, it can withstand high voltage while holding a low internal resistance.

FIG. 1

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FIELD OF THE INVENTION AND RELATED ART STATEMENT

1. FIELD OF THE INVENTION

5 The present invention relates generally to electric double layer capacitor and, in particular, it is concerned with such capacitors that use activated carbon in their polarizable electrodes and the method for producing the same.

DESCRIPTION OF THE RELATED ART

10

The electric double layer capacitor is a capacitor of large capacitance that uses activated carbon in its polarizable electrodes and utilizes a phenomenon that electric charge is stored in an electric double layer formed between the activated carbon and an electrolyte. Conventional electric double layer capacitors may roughly be classified into the following two categories: Namely, capacitors of a first group using electrolyte of aqueous solution such as aqueous sulfuric acid, and ones of a second group using electrolyte of organic solution wherein electrolyte is dissolved in an organic solvent such as propylene carbonate.

Representative arrangements of these capacitors of the first group and the second group are schematically shown in Figs. 4 and 5, respectively. The capacitor shown in Fig. 4 comprises a separator 41 sandwiched between two facing activated carbon powder electrodes 42, 43 being backed by two conductive electrodes 48, 40 whose fringes are abutted on annular packings of insulator rubber 44, 45. The activated carbon powder electrodes 42, 43 are prepared by compounding activated carbon powder with concentrated aqueous sulfuric acid which also serves as a binding agent for the activated carbon and by molding the prepared compound into pellets.

In contrast to this, the capacitors holding an electrolyte of an organic solvent have typical structure shown in Fig. 5. A paste consisting of activated carbon powder, a fluoride resin and methanol is extended on aluminum nets 50, 51 and dried to obtain activated carbon electrodes 52, 53 in a state of film which are then wound to a roll by interlaying a separator 54. The wound roll is thereafter impregnated with a mixed solution of propylene carbonate and tetraethylammonium perchlorate and then housed in a container. In this figure, numerals 55, 56, 57 and 58 are an anode lead wire, a cathode lead wire, a rubber cap and aluminum container, respectively.

The conventional capacitors holding the above-described two kinds of electrolytes have the following advantages and disadvantages, respectively. The advantages inherent to ones holding the aqueous electrolyte are low electric resistance and thus suited for a load of large current discharge while the disadvantages thereof are that it is able to withstand only a low voltage, say 1.0 V which inevitably depends on decomposition voltage of the electrolyte. If one wishes to use it in a high voltage load, the use of a large number of capacitors connected in series becomes unavoidable to create a problem of reliability in long service.

On the other hand, the capacitors of the organic solvent electrolyte are advantageous in their capability of withstanding relatively high voltage (about 3V) and thus are capable of being used in a circuit of higher voltage as compared with the ones of an aqueous electrolyte. The disadvantages are large internal resistance of the capacitors due to a high electric resistance of the electrolyte which is about 5 — 10 times as compared with that of the one of aqueous electrolyte and a difficulty in the use in a load of large electric current.

45 OBJECT AND SUMMARY OF THE INVENTION

It is therefore the primary object of the present invention to provide a capacitor which has both the advantageous features of the conventional capacitors of the aqueous electrolyte and of the organic solvent electrolyte. More particularly, it intends to realize a capacitor that withstands high voltage while holding a low internal resistance.

In accordance with the present invention which has the above-described object, there is provided an electric double layer capacitor which comprises a pair of polarizable electrodes, composed of electrically-conductive substrates coated with layers of a mixture of activated carbon with a water-soluble material-based binding agent, optionally including an agent for improving the electric conductivity of the layer, arranged so that the one faces with the other by being interposed with a separator, and an electrolyte.

In embodying the present invention, the electrically-conducting substrates may preferably be ones of plates, foils, nets, perforated plates or expanded metals of a valve action metal selected from the group consisting of aluminum, tantalum and titanium, whose surfaces may optionally be roughened by etching,

and may preferably have a thickness equal to or under 50 μm .

The layer composed of activated carbon and binding agent may preferably have a thickness equal to or under 100 μm , and may be formed on the surfaces of the substrate. The activated carbon may be in any form of powder or fiber. In the case of activated carbon fiber, it may also be usable in either form of woven cloth, unwoven fabric sheet, felt or calendared paper.

The binding agent may preferably be produced from a water-soluble material which may be dehydrolyzed to become insoluble after drying. The water-soluble material may be one of polysaccharides or their derivatives.

The polysaccharides or their derivatives may be one selected from the group consisting of starch, chitin, chitosan or their derivatives, methyl cellulose, cellulose derivatives having (a) carboxyl group(s) such as carboxymethyl cellulose and carboxyethyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, or their derivatives.

In the case of cellulose derivatives having (a) carboxyl group(s) such as carboxymethyl cellulose and carboxyethyl cellulose, they may preferably be one(s) at least one of whose proton in their carboxyl group is substituted by any of alkali metal ion, alkaline earth metal ion, ammonium ion or aluminum ion.

Alternatively, the binding agent may be one selected from the group consisting of polyvinyl pyrrolidone, polyvinyl alcohol and polyvinyl acetate, some of which may optionally be modified to be water-soluble for better dispersion in the manufacturing process.

The layers may further include an agent for improving the electric conductivity. The agent may be at least one selected from the group consisting of graphite, carbon black, acetylene black, ruthenium oxide and carbon fiber, and may preferably be powder having particle diameter equal to or below 1 μm .

According to another aspect of the present invention, there is also provided a method for producing the electric double-layer capacitor which comprises, depositing a mixture prepared by dispersing activated carbon powder and an agent for improving the electric conductivity of the layer, in an aqueous solution of methyl cellulose or carboxymethyl cellulose, on an aluminum substrate by either one means of spreading, impregnating and printing; drying the substrates with the layer of the deposited mixture; and then arranging the dried substrates so that the substrate face with each other by interlaying a separator therebetween.

In the stated method, either one of alcohol and ammonia may preferably be added at any of said dissolving, mixing or dispersing step.

Alternatively, the electric double layer capacitor of the present invention may be one which comprises; a polarizable electrodes composed of activated carbon and a binding agent, and having a volumetric filling factor equal to or above 0.4 (activated carbon/binding agent), an electrically-conductive electrode, separators, and an electrolyte. It is appreciated that the stated value (no less than 0.4) of the volumetric filling factor is preferred for optimum electric conductivity.

By embodying the present invention, the internal resistance of the obtained capacitor can be made extremely small because the invention provides an activated carbon electrode structure including a composition of a low electric resistance which gives, a property of excellent electrically-contacting with the electrically-conductive substrates as well as property of good film-forming and self-supporting to the activated carbon layer. Furthermore, since the thickness of the activated carbon layer of the activated carbon electrode composition of the present invention can be made extremely small, a capacitor of a large capacitance and of a low electrical resistance in view of geometrical space factor of the electrode (i.e. large surface area of the electrodes containable in a given unit volume, obtained by laminating a number of thin layers) is made readily available.

BRIEF EXPLANATION OF THE DRAWING

The foregoing and other characteristics of the present invention will be made clearer by the following detailed description of a preferred embodiment thereof given by way of non-limiting example, and by the annexed drawing, wherein:

Fig. 1 is a schematic perspective view, partly cut-out, of an example of a capacitor built in accordance with the present invention;

Figs. 2 and 3 are side cross-sectional views of the other embodiments, respectively; and

Figs. 4 and 5 are schematic views of conventional capacitors.

It will be recognized that some or all of the Figures are schematic representations for purposes of illustration and do not necessarily depict the actual relative sizes or locations of the elements shown.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

EXAMPLE 1

Ten parts by weight of activated carbon powder (specific surface area: 2000 m²/g, mean particle diameter: 2 μm) and 2 parts by weight of acetylene black were uniformly dispersed in a mixed solution of water and methanol. Apart from this, 2 parts by weight of carboxymethyl cellulose (CMC), whose protons in their carboxyl groups were partly substituted by Na ions, were dissolved in water. Both of the dispersion and the solution were then combined to be mixed together by stirring to give an activated carbon slurry. Thereafter, the obtained slurry was deposited by dipping on the both sides of an aluminum foil 1 (width: 10 mm, length: 40 mm, thickness: 20 μm), whose surface was roughened beforehand by chemical etching. After drying in the air for 30 min., the deposited foil was dried by applying far infra-red ray at 100°C for 80 min. to give activated carbon electrodes 2 and 3 having the superimposed layer of thickness: 50 μm. Incidentally, it was found that a favorable result was obtained if this thickness was equal to or under 100 μm.

A pair of the obtained foil electrodes 4 and 5 were wound interlaying a separator 6 therebetween. The wound body was then impregnated with an electrolyte prepared by dissolving tetraethylammonium tetrafluoroborate in propylene carbonate in 1 mol/l. Thereafter, the impregnated body was housed in an aluminum case 7 with aluminum lead wires 8 and 9, and a rubber packing 10, as illustrated by Fig. 1.

EXAMPLE 2

A capacitor, whose structure and composition were the same as that of EXAMPLE 1 except for omission of acetylene black, was prepared.

EXAMPLE 3

A capacitor, whose structure and composition were the same as that of EXAMPLE 1 except for the use of CMC whose protons in their carboxyl groups were partly substituted by NH₄ ions instead of Na ions, was prepared.

EXAMPLE 4

A capacitor, whose structure and composition were the same as that of EXAMPLE 1 except for the use of chopped phenolic resin-based activated carbon fiber (fiber diameter: 10 μm, mean chop length: 0.5 μm, specific surface area: 2300 m²/g) instead of the activated carbon powder, was prepared.

EXAMPLE 5

A capacitor, whose structure and composition were the same as that of EXAMPLE 1 except for the use of ruthenium oxide powder (mean particle diameter: 0.5 μm) instead of acetylene black, was prepared.

EXAMPLE 6

Ten parts by weight of activated carbon powder (specific surface area: 2000 m²/g, mean particle diameter: 2 μm) and 2 parts by weight of acetylene black were uniformly dispersed in aqueous ammonia (concentration: 5 % by weight). Apart from this, 2 parts by weight of carboxymethyl cellulose (CMC), whose protons in their carboxyl groups were partly substituted by Na ions, were dissolved in water. Both of the dispersion and the solution were then combined to be mixed together by stirring to give an activated carbon slurry. Thereafter, the obtained slurry was deposited by a roll coater on the both sides of an aluminum foil of thickness: 20 μm, whose surface was roughened beforehand by chemical etching. After drying in the air for 30 min., the deposited foil was dried by applying far infra-red ray at 100°C for 60 min. to give activated carbon electrodes.

A pair of the obtained foil electrodes were wound interlaying a separator therebetween to give an electrode body 20 shown in Fig. 2. The electrode body was then impregnated with an electrolyte prepared by dissolving tetraethylammonium tetrafluoroborate in propylene carbonate in 1 mol/l. And then the impregnated body was housed in a stainless steel case 21 with a stainless steel lid 23 having an aluminum layer 22 to complete a hermetically-sealed element. In the hermetically sealed element, both of positive/negative aluminum lead wires 24 and 25 were sealed with glass eyelets 26 and 27 which were strongly bonded to the lid 23, and the lid 23 was welded to the case 21 at their joint 28.

EXAMPLE 7

Ten parts by weight of activated carbon fiber chop (specific surface area: 2500 m²/g, mean length: 2 μ m, fiber diameter: 8 μ m) and 2 parts by weight of acetylene black were uniformly dispersed in a mixed solution of water and alcohol. Apart from this, 2 parts by weight of carboxymethyl cellulose (CMC), whose protons in their carboxyl groups were partly substituted by Na ions, were dissolved in water. Both of the dispersion and the solution were then combined to be mixed together by stirring to give an activated carbon slurry. Thereafter, the obtained slurry was deposited by printing method on one side of an aluminum foil 30 shown in Fig. 3 of thickness: 20 μ m, whose surface was roughened beforehand by chemical etching. After drying in the air for 30 min., the deposited foil was dried by applying far infra-red ray at 100° C for 60 min. to give activated carbon electrodes 31 and 32, which were then shaped to discs of diameter: 15 mm.

A pair of the obtained disc electrodes were combined together interlaying a separator 33 therebetween to give an electrode body which was then impregnated with an electrolyte prepared by dissolving tetraethylammonium tetrafluoroborate in propylene carbonate in 1 mol/l. Thereafter the impregnated body was housed in a pair of cases 34 and 35 with a gasket 36 to complete a capacitor of a coin type.

The ratings and other characteristics of the capacitors built in accordance with the above examples are determined as shown in TABLE below, in comparison with those (indicated by C.E. in the table) determined with the capacitors obtained by the comparative experiments.

TABLE

Invention:	Capacitance F or sec	Impedance		DC Resistance Ω	Reliability (-ΔC) %
		120 Hz	1KHz		
EXAMPLE 1	8 sec.	0.3	0.3	0.2	5
EXAMPLE 2	8.2 sec.	0.35	0.35	0.25	6
EXAMPLE 3	8 sec.	0.35	0.35	0.22	4
EXAMPLE 4	9 sec.	0.4	0.38	0.3	6
EXAMPLE 5	8 sec.	0.4	0.4	0.22	4
EXAMPLE 6	8 sec.	0.3	0.3	0.2	3
EXAMPLE 7	0.3 F	1.0	1.0	0.2	2
Prior Art:					
C.E. 1	4 sec.	2	1	2	30
C.E. 2	0.3 F	20	10	2	15
C.E. 3	6 sec.	0.3	0.3	0.2	40

The capacitor indicated by C.E. 1 in the above table is a wound one which has a layer (thickness: 200 μ m) composed of activated carbon and water-insoluble organic binding agent (fluorine containing synthetic resin), deposited on one side of an aluminum foil of thickness: 60 μ m, which is housed in a container of the same volume as that of EXAMPLE 1. The capacitor indicated by C.E. 2 is a coin type capacitor having a polarizable electrode composed of a woven cloth of activated carbon fiber. The capacitor indicated by of C.E. 3 is a capacitor with sulfuric acid electrolyte.

In the above table, the capacitances of EXAMPLES 1 -- 6 and C.E. 1 are indicated by the time period in second until the time points at which cell voltages reach 1 V at a discharging rate of 100 mA, and those of the others are indicated in Farad unit. The reliabilities are indicated by % of the decrease in the capacitance after the load test of 1.8 V for 1000 hours.

Although the present invention has been described in terms of the presently preferred embodiments, it is to be understood that such disclosure is not to be interpreted as limiting. Various alterations and modifications will no doubt become apparent to those skilled in the art after having read the above disclosure. Accordingly, it is intended that the appended claims be interpreted as covering all alterations and modifications as fall within the true spirit and scope of the invention.

Claims

1. An electric double layer capacitor which comprises;
a pair of polarizable electrodes, comprising of electrically-conductive substrates coated with layers
formed on said substrates, of a mixture of activated carbon and a water-soluble material-based binding agent,
separators interposed between said electrodes facing with each other, and
an electrolyte.
2. An electric double layer capacitor which comprises;
a pair of polarizable electrodes, comprising of electrically-conductive substrates coated with layers
formed on said substrates, of a mixture of activated carbon and at least one of polysaccharides or their derivatives which acts as a binding agent,
separators interposed between said electrodes facing with each other, and
an electrolyte.
3. An electric double layer capacitor which comprises;
a pair of polarizable electrodes, comprising of electrically-conductive substrates coated with layers
formed on said substrates, of a mixture of activated carbon and at least one binding agent selected
from the group consisting of methyl cellulose, cellulose derivatives having (a) carboxyl group(s) such as
carboxymethyl cellulose or carboxyethyl cellulose, polyvinyl pyrrolidone, hydroxyethyl cellulose, hydrox-
ypropyl cellulose, polyvinyl alcohol, polyvinyl acetate and their derivatives,
separators interposed between said electrodes facing with each other, and
an electrolyte.
4. The electric double layer capacitor as claimed in any one of Claims 1 --- 3, wherein said layers
superimposed on said electrically-conductive substrate further include an agent for improving electric
conductivity.
5. The electric double layer capacitor as claimed in any of Claims 1 -- 3, wherein said electrically-
conducting substrates are ones of plates, foils, nets, perforated plates, and expanded metals of a valve
action metal selected form the group consisting of aluminum, tantalum and titanium, whose surfaces
are optionally etched.
6. The electric double layer capacitor as claimed in any of Claims 1 --- 3, wherein said activated carbon is
in a form of powder or fiber.
7. The electric double layer capacitor as claimed in Claim 1, wherein said water-soluble material-based
binding agent is at least one selected from the group consisting of methyl cellulose, cellulose
derivatives having (a) carboxyl group(s) such as carboxymethyl cellulose or carboxyethyl cellulose,
polyvinyl pyrrolidone, hydroxyethyl cellulose, hydroxypropyl cellulose, polyvinyl alcohol, polyvinyl ace-
tate and their derivatives.
8. The electric double layer capacitor as claimed in Claim 7, wherein said cellulose derivatives having (a)
carboxyl group(s) are ones at least one of whose proton in their carboxyl group is substituted by any of
alkali metal ion, alkaline earth metal ion, ammonium ion or aluminum ion.
9. The electric double layer capacitor as claimed in Claim 2, wherein said polysaccharide is selected from

the group consisting of starch, chitin, chitosan or their derivatives.

10. The electric double layer capacitor as claimed in Claim 4, wherein said agent for improving electric conductivity is at least one selected from the group consisting of graphite, carbon black, acetylene black, rutenium oxide and carbon fiber.
11. The electric double layer capacitor as claimed in any of Claims 1 -- 3, wherein said electrically-conductive substrate has a thickness equal to or under 50 μm .
12. The electric double layer capacitor as claimed in any of Claims 1 -- 3, wherein said layer composed of activated carbon and binding agent has a thickness equal to or under 100 μm .
13. The electric double layer capacitor as claimed in any of Claims 1 -- 3, wherein said layer composed of activated carbon and binding agent is formed on either one or both of the surfaces of said substrate.
14. The electric double layer capacitor as claimed in Claim 10, wherein said agent for improving electric conductivity is powder having particle diameter equal to or below 1 μm .
15. A method for producing an electric double layer capacitor which comprises: depositing a mixture prepared by dispersing activated carbon in an aqueous solution of at least one of the group consisting of methyl cellulose, cellulose derivatives having (a) carboxyl group(s), polyvinyl pyrrolidone, hydroxyethyl cellulose, hydroxypropyl cellulose, polyvinyl alcohol, polyvinyl acetate and their derivatives, or in a solution prepared by dissolving polysaccharides in water or an aqueous acid, on aluminum substrates by either means of painting, impregnating, printing, spin coating, or doctor-blade coating; drying the substrates with the deposited layer of the mixture; and arranging the dried substrates with separators so that said substrates face with each other interposing separators therebetween.
16. The method for producing electric double layer capacitor as claimed in Claim 15, wherein said mixture prepared by the dispersion further includes an agent for improving electric conductivity.
17. The method for producing electric double layer capacitor as claimed in Claim 15, wherein at least either one of alcohol and ammonia is added at said dissolving, mixing or dispersing step.
18. The electric double layer capacitor as claimed in Claim 6, wherein said activated carbon fiber is in either form of woven cloth, unwoven fabric sheet, felt, chopped fiber or calendared paper.
19. An electric double layer capacitor which comprises; a polarizable electrodes composed of activated carbon and a binding agent, and having a volumetric filling factor equal to or above 0.4 (activated carbon/binding agent), an electrically-conductive electrode, separators, and an electrolyte.

FIG. 1

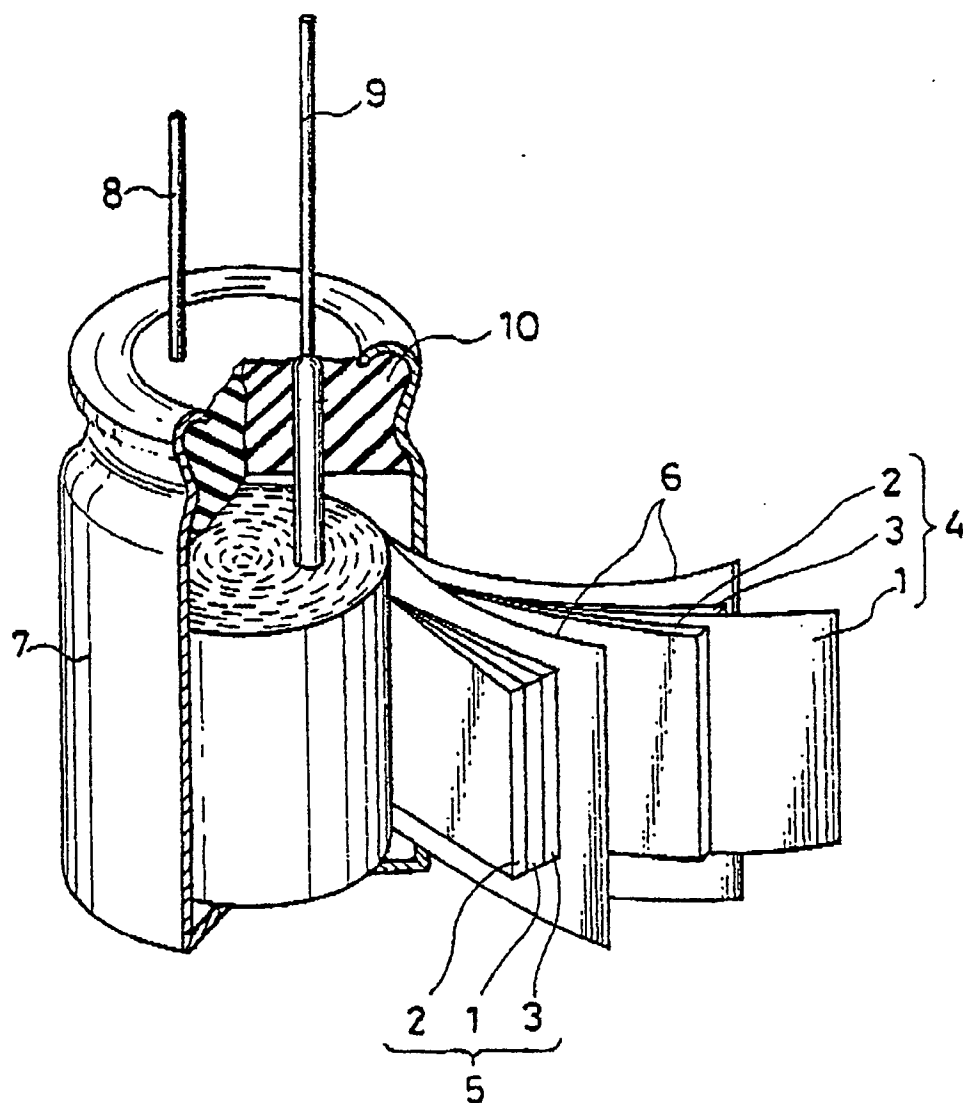


FIG. 2

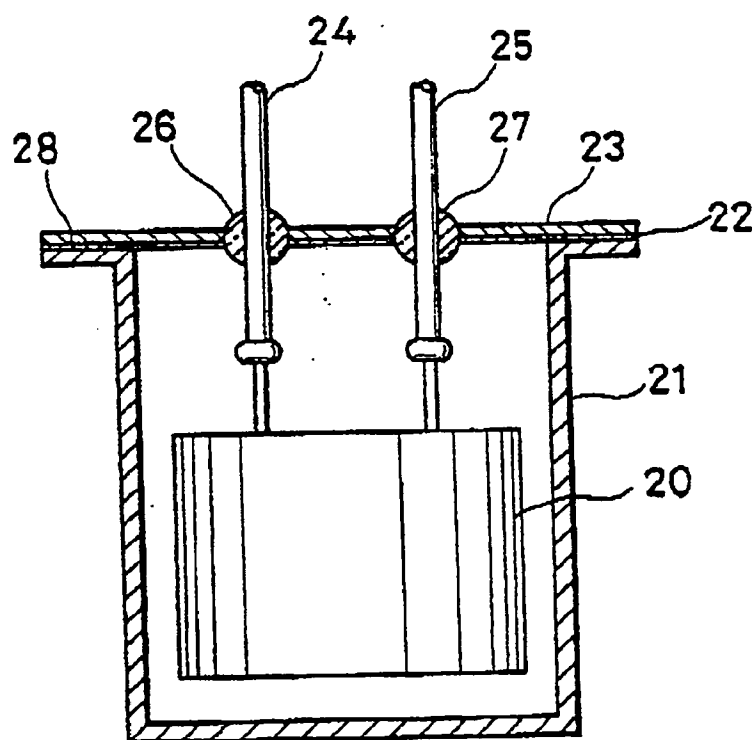


FIG. 3

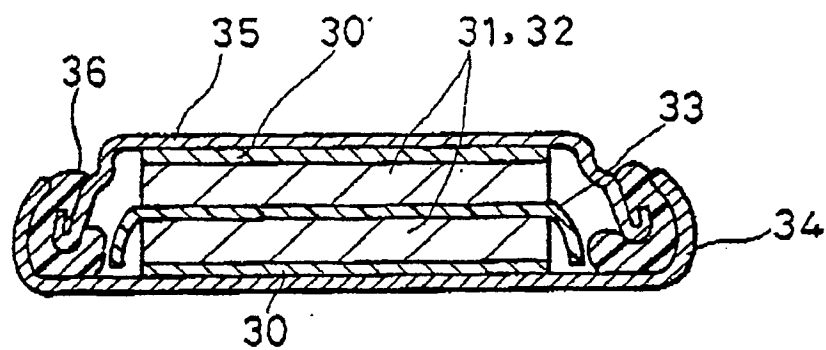


FIG. 4

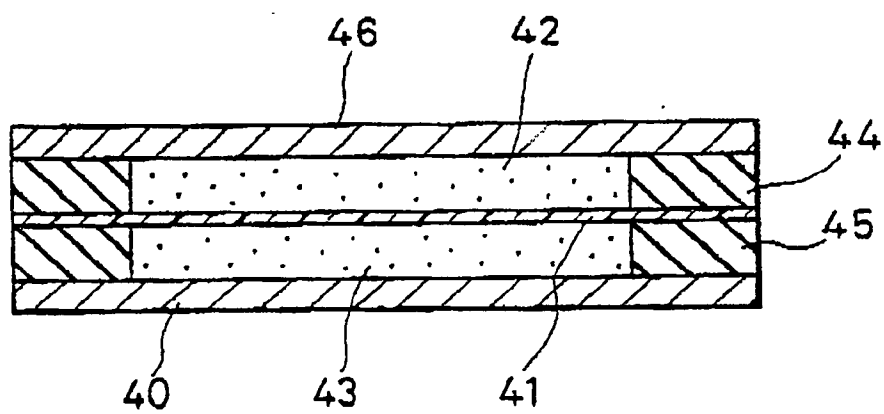
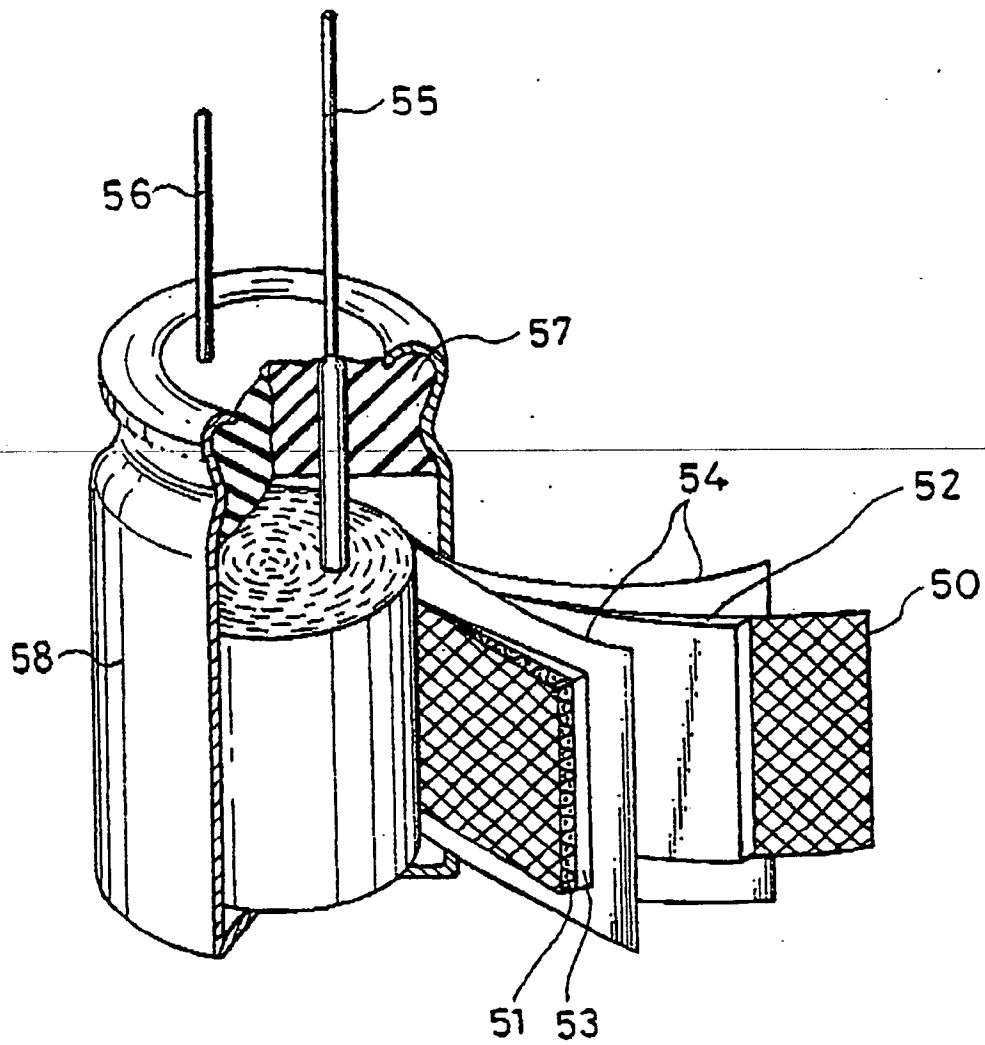


FIG. 5 (Prior Art)



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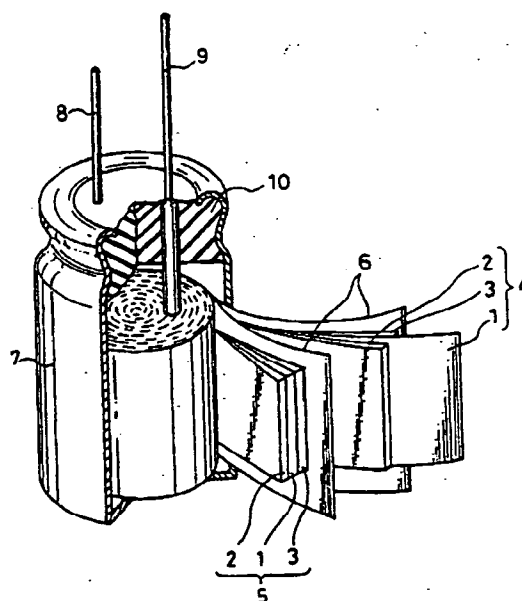
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(54) **Electric double layer capacitor and method for producing the same.**

(57) An electric double layer capacitor which comprises a pair of polarizable electrode (4, 5), composed of electrically-conductive substrates (1) coated with layers formed on said substrates, of a mixture of activated carbon with a water-soluble material-based binding agent, optionally including an agent for improving electric conductivity, facing with each other by being interposed with a separator (6), and an electrolyte is disclosed.

FIG. 1**EP 0 449 145 A3**



European
Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 91 10 4570

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X	GB-A-2 040 570 (MATSUSHITA ELECTRIC) * Abstract; page 1, lines 32-35,47-48; page 2, table 2, lines 32-33; page 3, lines 2-4; claims 1,3,4 *	1-19	H 01 G 9/00
A	EP-A-0 112 923 (MATSUSHITA ELECTRIC) * Page 1, lines 15-21; page 2, lines 2-3; page 3, lines 23-25; page 4, lines 4-17; page 7, lines 3-9; page 10, line 13; page 15, lines 21-24; page 18, lines 16-18; page 28, lines 19-21; claims 1,2,4,14,16,18; figure 8 *	5-7,10, 11,15,18	
A	EP-A-0 187 163 (MATSUSHITA ELECTRIC) * Page 1, lines 11-16; page 2, lines 15-19; page 3, line 26; page 4, lines 19-20; page 6, lines 20-22; page 9, lines 6-9; page 18, table 3; page 30, line 15; claim 34; figure 2 *	5,10,12, 14,15,18, 19	
A	DE-A-3 530 772 (MURATA MANUFACTURING) * Page 5, lines 16-20; page 6, lines 14-16; claim 2 *	15,17,19	
A	PATENT ABSTRACTS OF JAPAN, vol. 13, no. 352, 8th August 1989; & JP-A-01 112 719 (KURARAY) 01-05-1989 * The whole document *	8	TECHNICAL FIELDS SEARCHED (Int. Cl.5)
A	US-A-4 614 696 (ITO et al.) * Claims 3,4 *	1,3,7	H 01 G
A	WORLD PATENT INDEX LATEST, week 8227, accession no. 82-55778E, Derwent Publications Ltd, London, GB; & JP-A-57 085 027 (SEIKOSHA) 27-05-1982 * The whole document *	15	
The present search report has been drawn up for all claims			
Place of search A The Hague		Date of completion of search 16 October 91	Examiner GOOSSENS A.M.J.
CATEGORY OF CITED DOCUMENTS X: particularly relevant if taken alone Y: particularly relevant if combined with another document of the same category A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: earlier patent document, but published on, or after the filing date D: document cited in the application L: document cited for other reasons &: member of the same patent family, corresponding document			